

INVESTIGATION OF ADVANCED LIQUID LUBRICANTS

PART I - STUDY OF THE HIGH TEMPERATURE OXIDATIVE DEGRADATION
OF LIQUID LUBRICANTS

PART II - DEVELOPMENTAL SYNTHESIS OF HIGH TEMPERATURE THERMALLY
AND OXIDATIVELY STABLE LIQUID LUBRICANTS

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Air Force Materials Laboratory

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Project Nr 3044

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FOREWARD


This report was prepared by the Fluid and Lubricant Materials Branch, Nonmetallic Materials Division, Air Force Materials Laboratory, Aeronautical Systems Division. The investigation was initiated under Project No. 3044, "Aerospace Lubricants", Task No. 304405, "Liquid Lubricants", with Mr. Carl E. Snyder, Jr. acting as project engineer. The assistance of Mr. Mr. Lynn Applegate of the University of Dayton is gratefully acknowledged.

This report covers work conducted from 1 August 1962 to 10 May 1963.

ABSTRACT

This report covers work being done on the development of high temperature liquid lubricants for use in advanced aerospace applications. The phases of study include; (1) the selection of additives on a more scientific basis; and (2) modification of molecular structures of existing fluids by developmental synthesis. The work on Phase I covered two classes of materials, the alkyl-substituted pyrazines, and the aryl-esters. The work on Phase II was concerned with the modification of the polyphenyl ethers to improve their low temperatures properties by attacking various alkyl groups to the polyphenyl ether system through an amine linkage. This is an interim report, as work is continuing on these two phases.

This report has been reviewed and is approved.

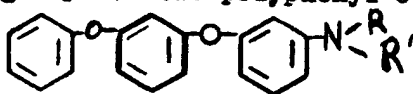

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I. INTRODUCTION

The current objective of subtask 304405A, the subtask under which the work covered in this report was performed, is to develop high temperature liquid lubricants for use in advanced aerospace applications. The current objective of this program is to develop an oxidatively stable fluid or formulation for use at bulk oil temperatures of 500° F and above. The objective is being pursued in two Phases.

Phase I. - The selection of additives on a mere scientific basis, incorporates the use of vapor phase chromatography, liquid-solid chromatography, infrared spectroscopy, and other research techniques to identify the oxidative degradation products of base fluids and various formulations of the base fluids under investigation. Upon the comparison of the oxidation products of the base fluid and its formulations with various additives, the effect of the additives on preventing the oxidation of the fluid is determined. After a back-log of information concerning the types of oxidation minimized by various additives is obtained, it is anticipated that the best additive package for a new experimental fluid could be predicted by studying its chemical structure.

Phase II. - The current emphasis on phase II, the modification of molecular structure of existing fluids by developmental synthesis, is on the improvement of the low temperature properties of the polyphenyl ethers. An attempt is being made to improve their low temperature properties by attacking various alkyl groups to the polyphenyl ether backbone through an amine linkage.

(i.e. ) It is hoped that the nitrogen atom being used as the link will sufficiently lower the electron density of the alkyl groups so that the entire system will be oxidatively stable at 500° F.

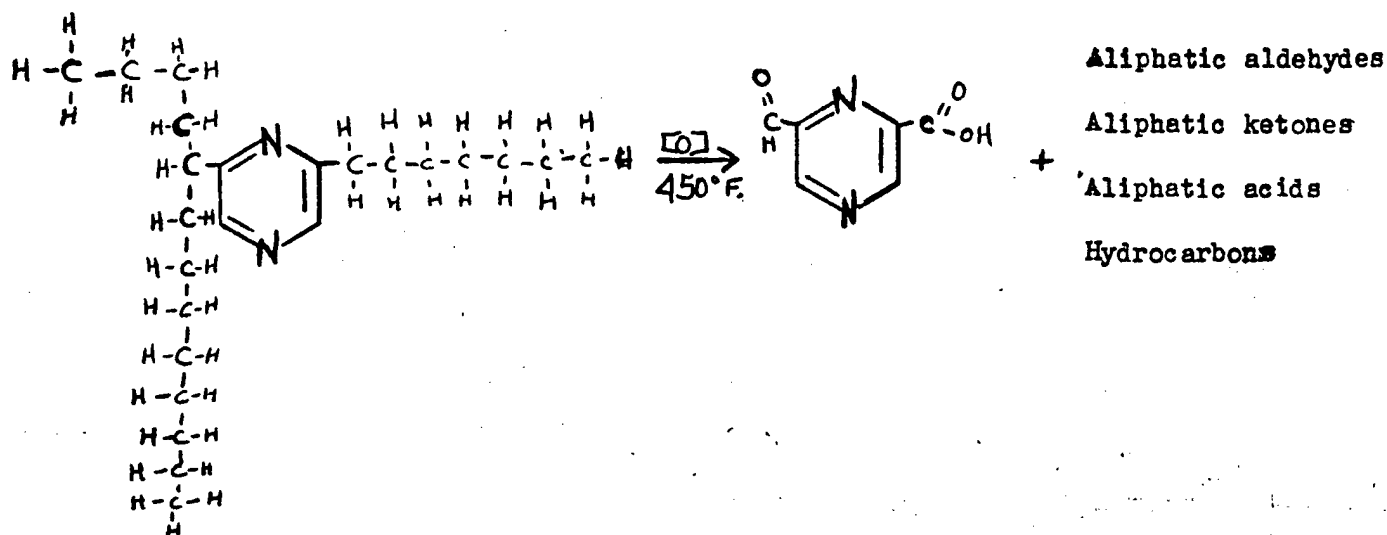
II. SELECTION OF ADDITIVES

In order to select an additive to inhibit the oxidative degradation of a specific fluid, it is first necessary to have a better knowledge concerning the oxidative degradation products. The following procedure is currently being used to obtain the needed information (a) a sample of an unformulated fluid is oxidatively stressed under specific conditions, with provisions made for separately collecting a dry-ice cooled condensate, a water-cooled condensate and a residue. (b) These three fractions are individually further separated and the components thereby obtained characterized and identified by vapor phase chromatography, infrared spectroscopy, and other research tools and techniques. (c) Samples of the unstressed base fluids are then formulated with various additives, the formulations oxidized as in (a), and the fractions thus collected separated and identified. Comparison of the products formed by the oxidation of the formulation with those formed in the base fluid, the effect of each additive can be determined.

Two fluids have been studied up to and including step (b). A disubstituted alkyl pyrazine fluid, was available only in minute quantities, thereby allowing only a preliminary investigation. However, a second fluid, an aryl ester, is available in sufficient quantity to carry the study through step (c), and this work has been recently initiated.

Although the supply of the alkyl pyrazine, 2-n-heptyl-6-(5-tridecyl)-pyrazine, was limited, some important information was obtained which confirmed predictions as to the mode of decomposition of this class of fluids. The fluid was oxidized at 450° F for 24 hours with an air flow of 20 l/hr, and three fractions were obtained, a dry-ice acetone trap condensate, a water cooled condensate, and a residue. Even though exact identification of oxidative degradation products was not possible in all cases, usually enough material was collected to indicate what type of compound it was (i.e., aliphatic ketone, aldehyde, acid, et cetera). From the data obtained,

the oxidative degradation of the alkyl pyrazines seems to proceed through cleavage of the alkyl groups, leaving the pyrazine ring intact. In the residue, however, a small amount of materials whose infrared spectrum resembled that of a phthalate was found, which would indicate rearrangement, if not oxidative attack, on the pyrazine ring. However, the major of the oxidative attack seems to be occurring on the alkyl groups according to the following reaction:

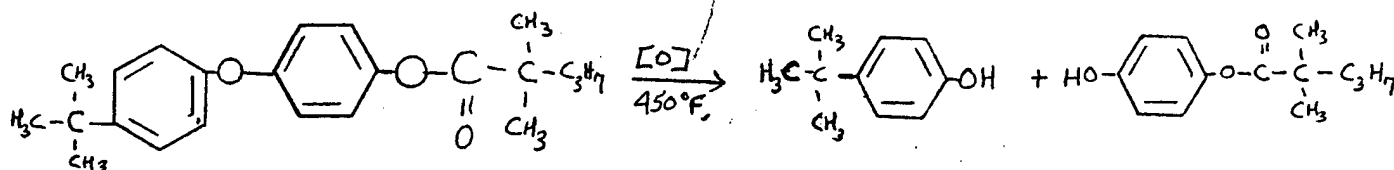


If more material would have been available, it would have been interesting to identify all of the aliphatic compounds formed, however it was not possible to obtain this information.

The work on the aryl-ester, was more complete because there was a sufficient quantity of the material under investigation, p-(tert-butylphenoxy)-phenyl neoheptanoate, to oxidatively stress enough samples for subsequent separation and identification of the oxidative degradation products. Only two samples were collected from the oxidation of this fluid, a water-cooled distillate, and a residue, as no dry-ice acetone cooled condensate was obtained. It was found that of the water-cooled distillate, the major component was neoheptanoic acid (72.5%). However, since the water-cooled distillate contained such a small quantity of

materials (3 ml), the residue contained the majority of the decomposition products. The major components of the residue were identified as (1) unoxidized starting material, (2) p-tert-butylphenol, (3) p-hydroxyphenylneheptanoate, and (4) hydrocarbons, probably alkenes (only a trace of this component was collected).

Since the major oxidative degradation products are p-t-butylphenol and p-hydroxyphenylneheptanoate, the site of the most vigorous oxidative attack seems to be the phenoxy-phenyl bond, (i.e.

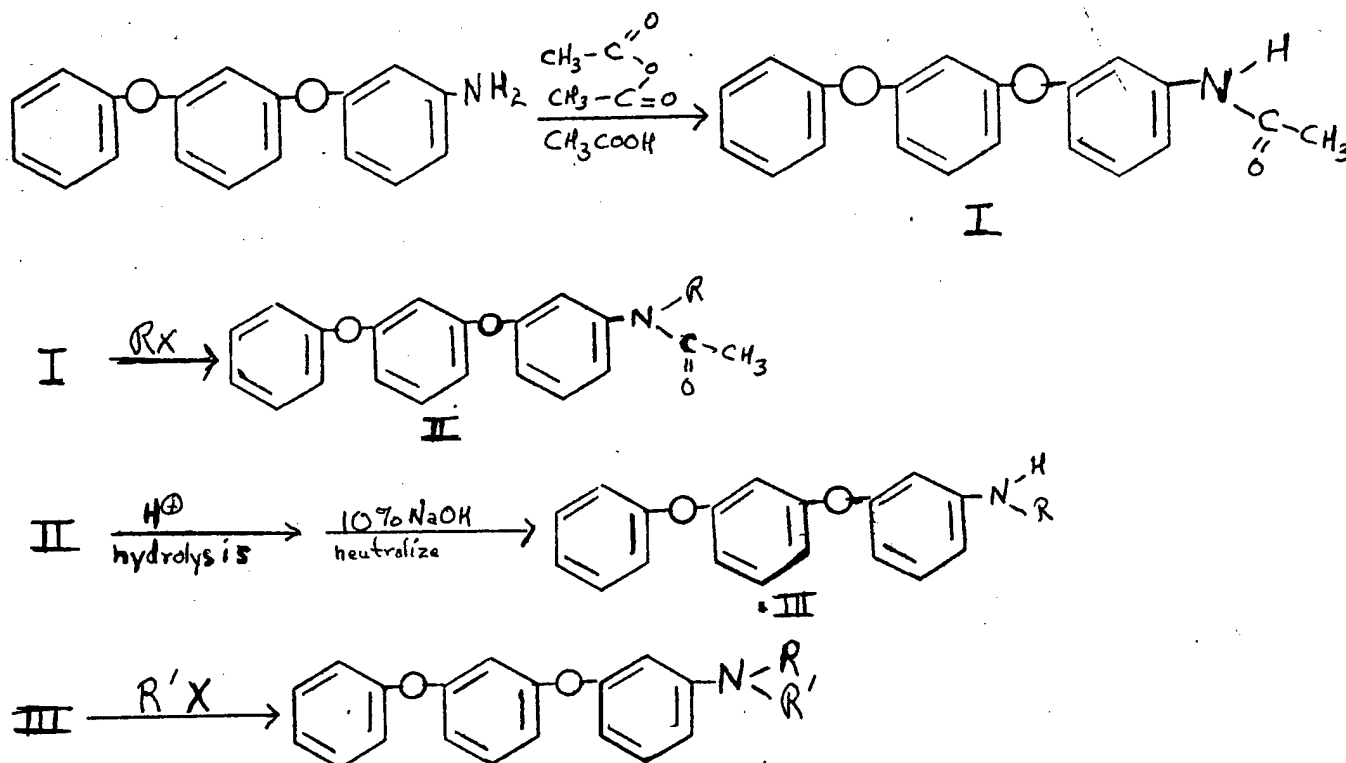


Whether this reaction proceeds through a peroxide or a furan ring intermediate has not yet been established, but since only compounds containing -OH groups were found, and no compounds containing mono-substituted benzene were detected, the peroxide intermediate seems to be favored. These data seem consistent with the findings that the same additives which are effective in the polyphenyl ethers, are effective in preventing the oxidative degradation of p-(tert-butylphenoxy)-phenyl neheptanoate. The effect of this additive on the oxidative degradation products of this fluid will be investigated by proceeding through Step C of this phase.

III. MODIFICATION OF MOLECULAR STRUCTURE BY DEVELOPMENTAL SYNTHESIS

Since the polyphenyl ether class of fluids exhibit excellent oxidative stability at 500° F, but possess poor fluid properties at low temperatures, a program was initiated to improve their low temperature properties without sacrificing their high temperature oxidative stability. This work supplements contractual effort being performed by Monsanto Research Corporation under contract AF 33(657)-8893.

From the work being done by Monsanto Research Corporation as well as by other similar contracts in which better low temperature properties were sought for various oxidatively stable systems by attaching alkyl groups directly to the oxidatively stable systems, it has been found that the inherent oxidative instability of the alkyl groups so decreases the oxidative stability of the whole system that the improved low temperature properties were of little value. However, it was felt that if the alkyl groups could be connected to the ring through an electronegative atom, which would change the electronic distribution throughout the alkyl groups, perhaps the alkyl groups would have greater oxidative stability. Therefore, the starting materials chosen for this synthetic effort was m-phenoxyphenyl-m-aminophenylether, and the alkyl groups would be linked to the polyphenylether system through an electronegative nitrogen atom by the following reaction sequence.



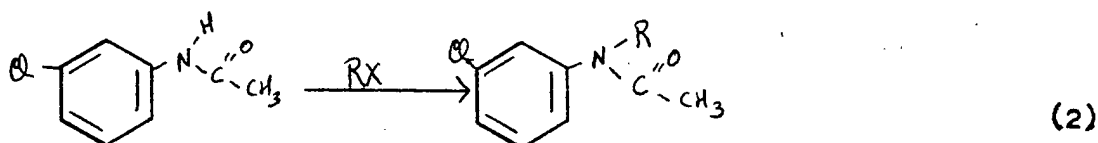
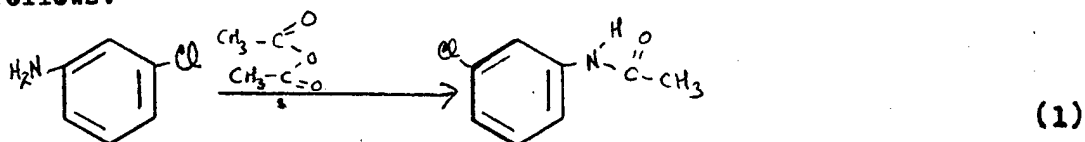
Where R and R' = alkyl groups (e.g. methyl, t-butyl, et cetera)

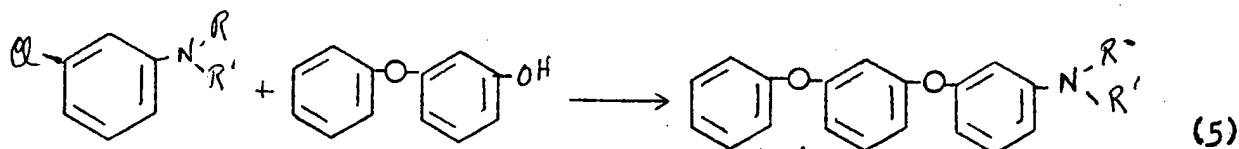
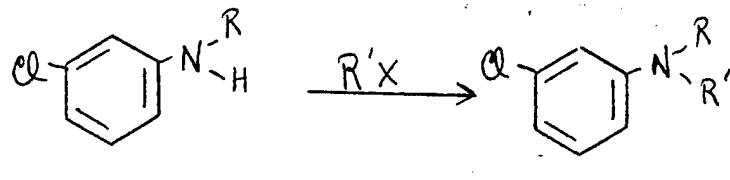
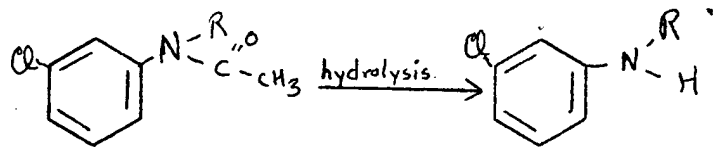
Acetylation of the starting material proceeded quite nicely, but alkylation of the acetylated amine has not proceeded nearly as smoothly. At first a small scale alkylation was attempted with 1-n-hexylbromide, and, although the reaction did occur, it went in such poor yield that it was impossible to calculate. Another alkylation was attempted with tert-butyl iodide and no vacuum distillable products were formed. Another procedure is being attempted to isolate and identify the reaction product.

IV. SUMMARY AND CONCLUSIONS

A. The results obtained in Phase I of this program, the selection of additives on a more scientific basis, indicate that vapor phase chromatography and/or liquid solid chromatography used in conjunction with infrared spectroscopy and other analytical methods show promise as research tools in the study of high temperature oxidative degradation products of fluids for use as liquid lubricants in advanced aerospace applications. Although sufficient quantity of the alkyl pyrazine studied was not available for a more complete investigation, the work was useful in the development of the techniques required for the program and showed that an approach of this type was feasible.

B. The work done on the chemical modification of the polyphenylethers indicates changes that need be made in the program. Due to the difficulty encountered in the isolation and purification of the N-acetyl-n-alkyl-m-phenoxyphenyl-m-amine phenyl ethers prepared thus far, perhaps a different set of reactions may simplify and perhaps eliminate the problems. The revised reaction sequence is as follows:





This reaction sequence will facilitate the purification of the intermediate amine and amide reaction products by permitting the purification of lower molecular weight compounds which require less strenuous temperature requirements for their purification than the higher molecular weight intermediates which are very difficult to isolate. By the time the molecule is required to withstand high temperatures in its isolation and purification, it should be able to withstand the required conditions.

Since results on both phases of the program are encouraging, it is recommended that this program be continued.

V. EXPERIMENTAL (304405-A-1)

A. Oxidative Degradation of ELO-62-96 2-n-heptyl-6-(5-tridecyl)-pyrazine

A test tube (500 mm X 22 mm), fitted with a take-off adaptor (which is fitted with a modified Dean-Stark trap and a gas inlet tube which extends to within 4 mm of the bottom of the test tube), was charged with 20 ml of ELO-62-96 2-n-heptyl-6-(5-tridecyl)-pyrazine. The test tube was heated at 450° F for 24 hours with a flow of 20 l/hr of air passing through the sample. During the course of the oxidation, some volatile materials were formed and collected in

the modified Dean-Stark trap (Sample 6296G). Some of the volatile materials which formed were not collected in the Dean-Stark trap, but passed on through and were collected in a dry ice/acetone trap (Sample 6296B). The material which remained in the test tube was collected (Sample 6296D).

1. Separation of components of Sample 6296C by Vapor Phase Chromatography

All of the vapor phase chromatography work done during the time covered by this report was performed on an F & M Model 500 Gas Chromatograph, employing helium as a carrier gas. The column employed for this separation was a 4 ft X 1/4 inch stainless steel column packed with 2.0 weight % Versamid 900 on 60 - 80 mesh Gas-Chrom P. Listed below are the instrument conditions employed for the separation:

Helium Bleed	-	25 ml/min.
Helium Flow	-	88 ml/min.
Column Temperature	-	programmed from 75 to 350° C at 11° C/min.
Detector Temperature	-	295° C
Injection Port Temperature	-	295° C
Sample Size	-	150 ul

A total of 10 components were detected and were in sufficient quantity for collection. The collection of the components was accomplished by connecting a 12.5 cm length of 3 mm glass tubing in a U-shaped configuration to the exhaust port of the detector, by means of a No. 500 Beckman Teflon adapter. The U-shaped tube was cooled by means of a liquid nitrogen bath to condense the fractions. Each sample was washed out of the collection tube with chloroform (Matheson, Coleman, and Bell, spectroquality grade). The "Chloroform wash" was deposited on small sodium chloride plates and heated to evaporate the chloroform. Infrared spectra of the samples were then recorded. The fractions collected, their elution temperatures, and interpretation of their infrared spectra are listed in table I.

TABLE I - Components of 6296C as Separated by Gas Chromatography

Sample	Elution Temperature (° C)	Remarks on Infrared Spectra of Samples
1	151-153	Weak spectrum, but C = O band was evident
2	153-156	Insufficient material for infrared spectrum
3	157-161	Indications of a ketone and an acid group
4	161-165	Insufficient material for infrared spectrum
5	166-170	All of these spectra were practically identical. In addition to the bands present in sample No. 3, bands were also present at 6.3, 6.5, and 9.8 μ . These three later bands also appear in the pyrazine compound before oxidation.
6	172-200	
7	200-210	
8	212-219	
9	229-240	
10	322-345	

There is evidence of a carboxylic acid and a ketone structure in all samples which were recorded in the infrared region. Samples 5 through 10 also have bands indicative of the pyrazine structure of the starting material. The first fractions may possibly be carboxylic acids and ketones of the side chains. The later fractions could possibly be a carboxylic acid and a ketone of the original material with the nitrogen ring intact.

2. Separation of Components of Sample 6296D by Liquid-Solid

Chromatography

A chromatography column (42 X 5.5 cm), packed with silica gel (50-200 mesh, chromatographic grade) was charged with 6296D (2.0g) dissolved in C Cl₄ (25 ml). The fractions collected are listed in Table II.

TABLE II - COMPONENTS OF 6296D AS SEPARATED BY LIQUID-SOLID CHROMATOGRAPHY

Fraction	Eluent	Volume	Appearance of solution
1	C Cl ₄	600 ml	Colorless
2	C Cl ₄	600 ml	Colorless
3	Benzene	250 ml	Colorless
4	Benzene	250 ml	Colorless
5	Chloroform	400 ml	Colorless
6	Chloroform	400 ml	Colorless
7	Chloroform	400 ml	Light Yellow
8	Chloroform	300 ml	Light Yellow
9	50% Chloroform 50% Ethyl Acetate	400 ml	Yellow
10	50% Chloroform 50% Ethyl Acetate	400 ml	Yellow
11	50% Chloroform 50% Ethyl Acetate	400 ml	Yellow
12	50% Chloroform 50% Ethyl Acetate	400 ml	Yellow
13	25% Chloroform 75% Ethyl Acetate	500 ml	Yellow
14	Methyl Alcohol	750 ml	Yellow
15	Methyl Alcohol	750 ml	Yellow

The fractions were evaporated to dryness, and then dissolved in a minimum amount of spectroquality solvent. Although a very small sample was present in most cases, fairly good infrared spectra were obtained after evaporating the solvent and depositing the sample on small sodium chloride plates. Most of the samples were contaminated with the silicone grease used to seal the glass joints of the chromatographic column. Although specific compounds could not be ascertained, the information which gained from the infrared spectra is tabulated in Table III.

TABLE III - Data Obtained from Infrared Spectra of Fractions of 6293D as Obtained by Liquid-Solid Chromatography

Fraction	Remarks
1	This spectrum resembled spectra of hydrocarbons. Silicone grease was evident from the spectrum (contamination).
2	This spectrum was similar to No. 1 with a slight amount of carbonyl present.
3	A carbonyl, possibly a carboxyl group, was evident with silicone grease contamination.
4	This spectrum had a strong carbonyl and other absorption bands indicative of a phthalate.
5,7,8,9&13	All of these spectra were practically identical to spectrum of fraction 4. Silicone grease was evident in all of these spectra, as a contaminant.
6&15	Strong evidence for a carboxylate ion.
10,11&12	The spectra were similar to the spectrum of the starting material 2-heptyl-6-(5 tridecyl)-pyrazine. A carbonyl was present in all three samples, also.
14	This spectrum gave evidence for a carbonyl, a carboxylate ion, and silicone grease contamination.

The mechanism by which a phthalate could be formed from the oxidation of the starting material can not be readily explained. Another sample of the starting material (ELO-62-96) should have been oxidized to determine the validity of the identification, but the supply of the material was exhausted.

3. Attempted Separation of Components of Sample 6296B

There was only enough sample for one run and the main component (80%) of this mixture was water.

B. Oxidative Degradation of ELO-62-93 p-(t-butyl phenoxy)-phenyl neoheptanoate .

A test tube (500 mm X 22 mm), fitted with a take-off adapter (which is fitted with a modified Dean-Stark trap and a gas inlet tube which extends to within 4 mm of the bottom of the test tube), was charged with 20 ml of ELO-62-93 p-(t-butyl phenoxy)-phenyl neoheptanoate . The test tube was heated at 450° F

for 48 hours with a flow of 20 l/hr of air passing through the sample. During the course of the oxidation, some volatile materials were formed and collected in the modified Dean-Stark trap (Sample 6293C). No materials were collected in a dry ice/acetone trap which was connected to the outlet of the Dean-Stark trap. The material that remained in the test tube was rinsed from the test tube with carbon tetrachloride (Sample 6293D).

1. Separation of Components of Sample 6293C by Vapor Phase Chromatography.

A 2 foot X 1/4 inch stainless steel column packed with 2.0 wt % silicone gum rubber on 60 - 80 mesh Chromasorb P. Listed below are the instrument conditions employed for the separation:

Helium Bleed: 25 ml/min

Helium Flow: 45 ml/min

Column Temperature: Programmed from 50 to 400° C at 7.9° C/min

Detector Temperature: 405° C

Injection Port Temperature: 350° C

Sample Size: 100 ul

A total of 10 components were detected, but only two (2) were of sufficient quantity to collect and identify. The components detected, their elution temperatures, and their relative percentage of the sample are tabulated in Table IV.

TABLE IV - Components of 6293C as separated by Gas Chromatography

Component	Elution Temperature (° C)	Relative % Volume
1	50	0.45
2	50-53	0.17
3	56-64	3.76
4	65-76	12.23
5	82-88	0.11
6	90-117	72.53
7	118-126	6.80
8	168-177	0.42
9	217-226	0.89
10	254-265	1.52

Only fractions 6 and 7 were collected in sufficient quantity for infrared spectra. The samples were washed out of the collection tube with spectroquality chloroform, transferred to sodium chloride plates, the chloroform evaporated, and the infrared spectra recorded. Fraction 6, accounting for 72.53% of the sample, was identified as neoheptanoic acid. Fraction 7 was identified as p-t-butyl-phenol.

2. Separation of Components of Sample 6293D by Liquid-Solid Chromatography

A chromatography column (42 X 5.5 cm), packed with activated alumina (20-200 mesh, chromatographic grade) was charged with 5.0 ml of the solution with 6293D in carbon tetrachloride. The fractions collected, and their respective eluents are listed in Table V.

Infrared spectra were taken of the various fractions by evaporating the eluent, dissolving the resulting residue in spectroquality chloroform, and running the residue dissolved in chloroform versus pure spectroquality chloroform in matched 0.50 mm sodium chloride sealed cells. Fractions 1 through 8 were identified

TABLE V

Fraction	Eluent	Volume (ml)
1	5% C Cl ₄ in ligroine	300
2	10% C Cl ₄ in ligroine	300
3	10% C Cl ₄ in ligroine	300
4	15% C Cl ₄ in ligroine	300
5	35% C Cl ₄ in ligroine	300
6	40% C Cl ₄ in ligroine	300
7	40% C Cl ₄ in ligroine	300
8	C Cl ₄	500
9	C Cl ₄	300
10	H C Cl ₃	500
11	H C Cl ₃	500
12	H C Cl ₃	300
13	H C Cl ₃	400
14	50% Ethyl Acetate in H C Cl ₃	500
15	Ethyl Acetate	400
16	Ethyl Acetate	300
17	Ethyl Acetate	300
18	Ethyl Acetate	300
19	Ethyl Acetate	300
20	20% Methanol in Ethyl Acetate	300
21	20% Methanol in Ethyl Acetate	300
22	20% Methanol in Ethyl Acetate	300

as hydrocarbons, and even though specific identification of each compound was not possible, their infrared spectra indicate that they are alkenes, with only minor differences in chemical structure. The next two (2) fractions were identified as unoxidized starting material (ELO-62-93). The next fraction (No. 16) which contained

enough material for identification yielded p-tert-butylphenol. Fraction 18 thru 20 contained p-hydroxyphenylbenzoate. Fractions 21 and 22 contained sodium acetate. At the present time, the origin of the sodium is not known, but is being investigated. Fractions not accounted for in the identification either were composed of mixtures of the identified products or did not contain sufficient sample for identification.

VI. EXPERIMENTAL (304405-A-2)

A. Reaction of m-phenoxyphenyl-m-amino-phenylether with Acetic Anhydride

A 250 ml three necked round bottom flask fitted with a Tsubore stirrer, Liebig condenser, and a 60 ml addition funnel with a pressure equilibrating arm fitted with a gas inlet tube, was charged with m-phenoxyphenyl-m-amino phenylether (20.0g, 0.072 mole) dissolved in glacial acetic acid (40 ml). The 60 ml addition funnel was charged with acetic anhydride (8 ml, 0.084 mole) in glacial acetic acid (20 ml). The apparatus was flushed with dry nitrogen and a positive nitrogen pressure was maintained during the course of the reaction through the use of a mercury bubbler connected to the condenser. The acetic anhydride solution was added dropwise, with stirring, to the reaction vessel. The reaction mixture was then heated and refluxed for 2-1/2 hours, cooled, and then poured into ice water. Tarry brown material which formed was recrystallized from ethanol-water, yielding white crystals (16.75g, 73% yield, m.p. 69.5 - 70.5° C). The material was submitted for analysis:

Calculated: C: 75.47%; H: 5.03%; O: 15.09; N: 4.40

Found: C: 75.43%; H: 5.49%; O: 15.49; N: 4.45

B. Reaction of N-Acetyl-m-phenoxyphenyl-m-amino-phenylether with 1-bromohexane

Prior to assembling the apparatus, was baked dry overnight. A 100 ml three necked round bottom flask, fitted with a Tru-bore stirrer, a reflux condenser, and a Claissen adapter to which two 60 ml addition funnels were fitted (one fitted with a gas inlet tube), was charged with NaH (0.3g, 0.01 mole, in 25% mineral oil) in 35 ml of xylene after the apparatus had been purged with dry nitrogen. A dry

nitrogen atmosphere was maintained during the course of the reaction. One addition funnel was charged with N-acetyl-m-phenoxyphenyl-m-amino-phenylether (3.2g, 0.01 mole) in 25 ml of xylene. The other addition funnel was charged with 1-bromohexane (1.4 ml, 1.64g, 0.01 mole) in 15 ml of xylene. The N-acetyl-m-phenoxyphenyl-m-amino phenylether solution was added to the NaH slurry dropwise with stirring. The reaction mixture was then heated to reflux with stirring for two hours. During the course of the refluxing, the appearance of the reaction mixture changed from a milky gray to a clear yellow solution. At the completion of two hours of refluxing, the 1-bromohexane solution was added dropwise, with stirring to the reaction flask. After the dropwise addition of the 1-bromohexane solution was completed, the reaction mixture was heated and refluxed for 1/2 hour, during which time the appearance of the reaction changed from a clear yellow to a milky-white solution. The reaction mixture was cooled to room temperature and filtered. The white solid which was collected gave a positive halogen test when tested with Ag NO₃ solution. The filtrate was washed twice with water and the xylene was evaporated, yielding a white oil. The material failed to crystallize from ethanol-water, so it was vacuum distilled at a bath temperature of 320° C. A small quantity of a colorless liquid (b.p. 208° C/0.085 mm Hg) was collected and an infrared spectrum of this materials indicated that the reaction had yielded a trace of N-n-hexyl-N-acetyl-m-phenoxyphenyl-m-amino-phenylether. No other materials were isolatable from the tar which had formed during the vacuum distillation. Perhaps the use of nitrogen through the capillary bleed during the vacuum distillation would result in less tar formation.

C. Reaction of N-acetyl-m-phenoxyphenyl-m-amino-phenylether with t-butyl Iodide

Prior to assembling the apparatus was baked dry overnight. A 500 ml three necked round bottom flask, fitted a Tru-bore stirrer, reflux condenser, and Claissen adapter to which two 100 ml addition funnels (one was fitted with a gas inlet tube) were fitted, was charged with NaH (1.5g, 0.05 mole, in 25% mineral oil) in 100 ml xylene

after the apparatus had been purged with dry nitrogen. A dry nitrogen atmosphere was maintained during the course of the reaction. One addition funnel was charged with N-acetyl-m-phenoxyphenyl-m-aminophenylether (16.0g, 0.05 mole) in 90 ml xylene. The N-acetyl-m-phenoxyphenyl-m-aminophenylether solution was added to the NaH slurry dropwise with stirring. The reaction mixture was then heated to reflux with stirring for two hours. During the course of the refluxing, the appearance of the reaction changed from a milky gray to a clear yellow solution. Then the t-butyl iodide solution was added dropwise with stirring to the reaction flask. After approximately 15 ml of the t-butyl iodide solution was added, fine white solids began to form. After the dropwise addition of the t-butyl iodide solution was completed, the reaction mixture was heated at reflux for 1/2 hour, the end of which time the reaction mixture changed from a clear yellow to a milky-white appearance. The reaction mixture was cooled to room temperature and filtered. An attempt was made to vacuum distill the filtrate, but after the xylene was stripped off, the bath temperature was raised to 370° C, and at a vacuum of 0.4 mm Hg, but no product was obtained. The reaction product is currently being worked up in a different manner, in an attempt to isolate the desired product.